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Chemodiversity of exudate flavonoids in some members of the Lamiaceae

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Abstract

Several newly studied species and further accessions of the Lamiaceae have been analyzed for their exudate flavonoid profiles. The principal compounds accumulated were flavones and their 6-methoxy derivatives, whereas flavonols were rarely encountered. The chemodiversity observed was relatively low, with only some 15 derivatives being found. The new data are discussed in relation to published data, and chemosystematic aspects are briefly addressed. Of the studied species, *Salvia arizonica* yielded only a rare diterpene quinone, demethylfruticulin A. Glandular hair diversification and different qualities of their secretions are briefly discussed.

Keywords: *Teucrium*; *Salvia*; *Phlomis*; *Dorystoechas*; Lamiaceae; Exudate flavonoids; Diterpene quinone; Chemodiversity; Chemosystematics

1. Introduction

The family of Lamiaceae consists of approximately 200 genera of cosmopolitan distribution, many of them of economic importance due to essential oil production. Most genera of the Lamiaceae are thus rich sources of terpenoids, but in addition a variety of iridoid glycosides and flavonoids is accumulated in considerable amount

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(Tomás-Barberán and Gil, 1990, 1992). Particularly the accumulation of iridoids in Lamiaceae and related families proved to be of chemosystematic relevance (Grayer et al., 1999). Within the flavonoids, 6/8-substituted flavonoids, predominantly flavone derivatives, are quite often encountered in this family (Tomás-Barberán and Wollenweber, 1990; Tomás-Barberán et al., 1988a, b; Grayer et al., 1999). Earlier, a strong tendency towards further A-ring substitution had been observed, and the frequencies of substitution patterns discussed (Wollenweber and Jay, 1988).

General surveys on flavonoid composition include distribution studies on exudate flavonoids within the family (Tomás-Barberán and Wollenweber, 1990) and rather rare comparative studies in single genera or their parts (*Thymus*: Hernandez et al., 1987; *Teucrium*: Harborne et al., 1986; *Sideritis*: Tomás-Barberán et al., 1988a, b; *Ocimum*: Grayer et al., 1999). In fact, some 16 genera of this family yielded no exudate flavonoids at all (Tomás-Barberán and Wollenweber, 1990). Similarly, within single genera several species or species groups were often reported to be devoid of exudate flavonoid constituents (Tomás-Barberán et al., 1988a, b). It is thus not surprising that infrageneric chemosystematic studies concentrate rather on flavonoid glycosides (e.g. Grayer et al., 2002; Upson et al., 2000). General polyphenol distribution within *Salvia*, covering a range of compounds including flavonoids, was recently surveyed (Lu and Foo, 2002).

In the present study, the exudate flavonoid profiles of some additional species of *Teucrium*, *Salvia*, *Dorystoechas* and *Phlomis* were studied for chemodiversity. In this context, chemodiversity[1] is understood as the expression of substitution profiles (in the sense of chemical molecular variation) on the basis of one defined molecular structure of known biosynthetic origin. The degree of chemodiversity and chemosystematic aspects are discussed, and results are compared with published data.

2. Materials and methods

Plant material was either collected from natural habitats or cultivated in the Botanical Gardens of the Technical University Darmstadt and of the University of Vienna, respectively. If not indicated otherwise, voucher specimens are deposited in the Herbarium of the Institute of Botany, University of Vienna (WU; see also Table 1). Aerial plant material, either fresh or thoroughly air-dried, was briefly rinsed with acetone to avoid extraction of leaf tissue constituents. The residues obtained after evaporation of the solvent were in most cases analyzed directly by TLC. Where bulk material was used, we proceeded as reported previously; i.e. terpenoids were eliminated by passing methanolic solutions of the exudates through Sephadex LH-20 and the flavonoid portions were further chromatographed on polyamide SC-6 (see e.g. Wollenweber et al., 2000). Flavonoid aglycones were identified by direct TLC comparison with markers available in E.W.'s laboratory. The identity of the diterpene quinone demethylfruticulin A (1; Fig. 1) isolated from *Salvia arizonica* was con-

[1] A theoretical paper on this subject is currently being prepared.

Table 1
Plant sources and geographical distribution

Name	Source of material	Geographic distribution and habitat information[a]
Dorystoechas hastata Boiss. & Heldr.	Cultivated, Botanical Garden, TU Darmstadt, coll. 2000, 3753, WU	Turkey: endemic to Southwest Anatolia: among rocks, also in Phrygana
Phlomis fruticosa L.	Greece, Prov. Ionannina, leg. K + H. Vetschera, Aug. 1985, WU	Eastern Mediterranean: dry places in mountain regions
Salvia arizonica Gray	Cultivated, Botanical Garden, TU Darmstadt, coll. 2000, 2298, WU	USA, Arizona: canyons, open forests and rocky soils (mountains of Trans-Pecos)
Salvia candelabrum Boiss.	Cultivated, Botanical Garden, TU Darmstadt, coll. 2000, 3529, WU	Western Mediterranean (Spain): dry places in submontane regions
Salvia dominica L.	Greece, Island of Crete, coll. Schneckenburger 06/2001, WU	Eastern Mediterranean: endemic; chalky hills
Salvia sclarea L.	Cultivated, Botanical Garden, TU Darmstadt, coll. 2000, 1060, WU	Southern Europe, Southwest and Central Asia: rocky igneous slopes, fields, road sides
Salvia syriaca L.	Iran, coll. Rustaiyan 1993, KAR	Western Syria, Armenia, Northern Iraq, Syrian desert, Iran. Steppe, fallow fields
Tecurium arduini L.	Cultivated, Botanical Garden, University of Vienna, LAM98E/008, WU	Western Yugoslavia, Northern Albania: rocky places
Teucrium botrys L.	Cultivated, Botanical Garden, University of Vienna, LAM98E/001, WU	South, West and Central Europe: dry, stony places
Teucrium canadense L.	Cultivated, Botanical Garden, University of Vienna, LAM98E/009, WU	North America: moist, wet soils
Teucrium chamaedrys L. (1)	Cultivated, Botanical Garden, University of Vienna, LAM98E/002, WU	Europe: open forests, cliffs, steppe
Teucrium chamaedrys (2)	Cultivated, Botanical Garden, University of Vienna, LAM98E/017, WU	Europe: open forests, cliffs, steppe
Teucrium hircanicum L.	Cultivated, Botanical Garden, University of Vienna, LAM98E/010, WU	Iran, Caucasus, East Anatolia: moist places
Teucrium marum L.	Cultivated, Botanical Garden, University of Vienna, LAM 98E/23, WU	Western Mediterranean (islands): dry, stony places
Teucrium puechiae Greuter ex Burdet	Cultivated, Botanical Garden, University of Vienna, LAM98E/24, WU	Southern Europe, Turkey: dry places
Teucrium scorodonia L.	Germany, woods near Darmstadt. Wollenweber, 2000, 6266, WU	Southern, Western, Central Europe: forests

[a] Distribution/habitat data taken from: Correll and Johnston (1970); Davis (1982); Gleason and Cronquist (1991); Tutin et al. (1972).

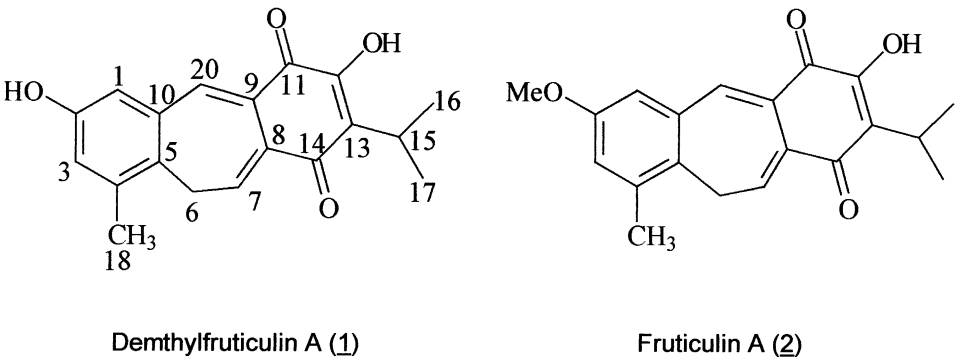

Fig. 1. Formulae of diterpene quinones

firmed by its NMR spectra, in comparison to those of fruticulin A (2 in Fig. 1; Rodriguez-Hahn et al., 1989): ^{1}H NMR (400 MHz, DMSO-d_6): δ6.89 (1H, d, J = 2.4 Hz; H-1), 6.85 (1H, d, J = 2.4 Hz; H-3), 3.04 (2H, d, J = 7.6 Hz; H-6), 6.82 (1H, t, J = 7.6 Hz; H-7), 7.95 (1H, s; H-20), 2.37 (3H, s; H-18), 3.27 (1H, m; H-15), 1.18 (6 H, d, J = 7 Hz; H-16/17), 9.45 (1H, s; OH), 10.67 (1H, br s; OH).

^{13}C NMR (100 MHz, DMSO-d_6): δ 114.0 (C-1), 156.6 (C-2), 121.6 (C-3), 135.6 (C-4), 130.1 (C-5), 27.6 (C-6), 139.2 (C-7), 132.5 (C-8), 130.9 (C-9), 135.4 (C-10), 183.6 (C-11), 154.9 (C-12), 125.2 (C-13), 182.5 (C-14), 24.1 (C-15), 19.4 (C-16/17), 19.6 (C-18), 133.2 (C-20).

3. Results

The species studied now belong to different subfamilies of the Lamiaceae such as the Teucrioideae (*Teucrium*), Nepetoideae (*Salvia*, *Dorystoechas*) and Lamioideae (*Phlomis*). Currently, there are eight recognized subfamilies with the largest, Nepetoideae, being further subdivided into tribes. This recently proposed classification was substantially corroborated by studies of the distribution of phenolic compounds in a large number of genera (Pedersen, 2000).

All accessions and taxa now studied accumulated mainly flavone derivatives in their exudates with a tendency towards 6-methylation. Flavonols were rarely encountered. Plant sources and geographic distribution are presented in Table 1, and results are presented in Tables 2 and 3, respectively.

3.1. Chemodiversity in Teucrium spp.

The genus *Teucrium* comprises some 115 species grouped in six sections and several subsections (Kästner, 1989). They occur in various habitats, including both very dry and moist places (Table 1). Systematically, some groups such as the species around *T. polium* still need revision. In general, the species and accessions of *Teucrium* studied exhibited rather low amounts of exudate flavonoids, except for *T.*

Table 2
Exudate flavonoids of *Teucrium* spp.

Teucrium	Apigenin	Ap-7-OMe	Ap 7,4′-diOMe	Scut 6,7 diMe	Luteolin	Lut 7-OMe	Lut 7,3′-diOMe	Lut-7,3′,4′-triOMe	6-OH-Lut-6,7-diOMe	6-OH-Lut-6,7,3′-triOMe	6-OH-Lut-6,7,3′,4′-tetraOMe
marum/CH–M			■		■	■	■		■	■	■
marum[a]				■	+				+	■	■
puechiae = *polium* s.l./CH-PO		■	■						■		
polium[a]/CH-PO				■	+				■	+	
chamaedrys/CH-CH (1)									■		
chamaedrys (2)											
chamaedrys[a]				■	+				■		
scorodonia/SC-SC					■				■		
scorodonia[a]				■	+				■		
canadense/SC-C					■				■		
arduini/SC-C					■				■		
arduini[a]				■	+				■		
hircanicum/SC-ST									+		
botrys/SC-B	■	■	■					■	■		
botrys[a]					+				■		

Abbreviations of compounds listed incl. semisystematic and trivial names

Compound (trivial name)	Semisystematic name
Apigenin	5,7,4′-triOH flavone
Ap-7-OMe (Genkwanin)	5,4′-diOH-7-OMe flavone
Ap 7,4′-diOMe	5-OH-7,4′-diOMe flavone
Scut 6,7 diMe (cirsimaritin)	5,4′-diOH-6,7-diOMe flavone
Luteolin	5,7,3′,4′-tetraOH flavone
Lut 7-OMe	5,3′,4′-triOH-7-OMe flavone
Lut 7,3′-diOMe (velutin)	5,4′-diOH-7,3′-diOMe flavone
Lut-7,3′,4′-triOMe	5-OH-7,3′,4′-triOMe flavone
6-OH-Lut-6,7-diOMe (cirsiliol)	5,3′,4′-triOH-6,7-diOMe flavone
6-OH-Lut-6,7,3′-triOMe (cirsilineol)	5,4′-diOH-6,7,3′-triOMe flavone
6-OH-Lut-6,7,3′,4′-tetraOMe	5-OH-6,7,3′,4′-tetraOMe flavone

Abbreviations referring to sectional groupings (Kästner, 1989): CH - M=sect. *Chamaedrys* subsect. *Marum*; CH - PO = sect. *Chamaedrys* subsect. *Polium*; CH - CH = sect. *Chamaedrys* subsect. *Chamaedrys*; SC - SC = sect. *Scorodonia* subsect. *Scorodonia*; SC - C=sect. *Scorodonia* subsect. *Canadensia*; SC - ST = sect. *Scorodonia* subsect. *Stachyobotrys*; SC - B=sect. *Scorodonia* subsect. *Botrys*■, Major compound; +, trace compound.(1), (2) refer to sources from Table 1.

[a] Literature data taken from Harborne et al. (1986).

Table 3
Exudate flavonoids of *Salvia* spp. and *Dorystoechas hastata*

Salvia	Apigenin	Ap-7-OMe	Ap 7,4′-diOMe	scut 7,4′-diMe	Scut 6,7-diOMe	Scut 6,7,4′-triOMe	Luteolin	Lut 3′-OMe	Lut- 7-OMe	Lut 7,3′-diOMe	6-OH-Lut 6,7-diOMe	6-OH-Lut 6,7,4′-triOMe	6-OH-Lut 6,3′,4′-triOMe	6-OH-Lut 6,7,3′,4′ tetraMe	Kae 3-OMe	Kae 3,7,4′-triOMe
candelabrum	■						■									
candelabrum[a]	■	■				■	■									
sclarea	■	■	■		■				■		■		■			
sclarea[a]	■	+	■			■	■									
syriaca		■	■		■	■				■		■				
syriaca[b]			■	■		■						■		■		
dominica	■				■	■	■	■			■			■	■	
arizonica	No flavonoids detected; diterpene quinones only															
Dorystoechas hastate			■													■

Abbreviations of compounds listed incl. semisystematic and trivial names

Compound (trivial name)	Semisystematic name
Apigenin	5,7,4′-triOH flavone
Ap-7-OMe (Genkwanin)	5,4′-diOH-7-OMe flavone
Ap 7,4′-diOMe	5-OH-7,4′-diOMe flavone
Scut 7,4′-diMe (ladanein)	5,6-diOH-7,4′-diOMe flavone
Scut 6,7 diMe (cirsimaritin)	5,4′-diOH-6,7-diOMe flavone
Scut 6,7,4′-triOMe (salvigenin)	5′-OH-6,7,4′-triOMe flavone
Luteolin	5,7,3′,4′-tetraOH flavone
Lut 3′-OMe (chrysoeriol)	5,7,4′-triOH-3′-OMe flavone
Lut 7-OMe	5,3′,4′-triOH 7-OMe flavone
Lut 7,3′-diOMe (velutin)	5,4′-diOH-7,3′-diOMe flavone
6-OH-Lut-6,7-diOMe (cirsiliol)	5,3′,4′-triOH-6,7-diOMe flavone
6-OH-Lut 6,7,4′-triOMe (eupatorin)	5, 3′-diOH-6,7,4′-triOMe flavone
6-OH-Lut 6,3′,4′-triOMe (eupatilin)	5, 7-diOH-6,3′,4′-triOMe flavone
6-OH-Lut-6,7,3′,4′-tetraOMe	5-OH-6,7,3′,4′-tetraOMe flavone
Kae 3-OMe (isokaempferide)	5,7,4′-triOH-3-OMe-flavone
Kae 3,7,4′-triOMe	5-OH-3,7,4′-triOMe flavone

■, Major compound; +, trace compound.
[a] Literature data taken from Adzet et al. (1988).
[b] Literature data taken from Hatam and Yousif (1992).

marum, which is more aromatic than the other taxa. Similarly, the chemodiversity of exudate aglycones is rather low. Cirsiliol (6-hydroxyluteolin 6,7-dimethyl ether) occurs throughout the range of species. 6-Methoxylation appears to be restricted to derivatives of luteolin; its derivatives dominate over those of apigenin (Table 2). However, literature reports on exudate flavonoids of some European species indicate the presence of corresponding 6-methoxyapigenin derivatives (Harborne et al., 1986; sub (a) in Table 2). Inclusion of these data permits comparison of different accessions. The results largely coincide with the exception of cirsimaritin (scutellarein 6,7-dimethylether), which was presently not found in any *Teucrium* species. By and large the exudate profiles appear to be quite stable at the infraspecific level.

Further literature data refer mainly to extracts and are thus not fully comparable to exudate results. Cirsimaritin (scutellarein 6,7-diOMe), eupatorin (6-OH-luteolin 6,7,4′-trimethyl ether), apigenin 7,4′-diOMe and cirsiliol (6-OH-luteolin 6,7-dimethyl ether) have been isolated from leaf extracts of *T. polium* L., a near relative to *T. puechieae* (Verykokidou-Vitsaropoulou and Vaijas, 1986). Whole plant extracts yielded salvigenin (6-OH apigenin 6,7,4′-trimethyl ether) and cirsiliol (Rizk et al., 1986). Kawashty et al. (1999) reported trace amounts of cirsimaritin as a leaf/stem constituent from the same species. Extracts of aerial parts of *T. hircanicum* yielded pedalitin (6-hydroxyluteolin 7-methyl ether; Oganesyan and Mnatsakanyan, 1987), while cirsimaritin was found as a free aglycone in extracts of *T. arduini* (Kalodera et al., 1993).

Contrary to the frequently observed correlations between exudate flavonoid accumulation and xeric or alpine habitats (e.g. Wollenweber et al., 1996), species such as *T. canadense* (moist habitats) and *T. arduini* (dry habitats; Table 1) yielded identical exudates, both quantitatively and qualitatively. Of all species studied here, *T. marum* is the most aromatic species, and indeed exhibited the highest yield and degree of chemodiversity. In this case, correlation to xeric habitats is evident. The chemosystematic value of the observed chemodiversity of exudate flavonoids is still obscure, despite earlier studies indicating singular compounds (salvigenin) as markers at the sectional level. From our results, only *T. marum* and *T. botrys* differ significantly from the rest of the analyzed species, but neither a specific profile nor a single compound appears to be characteristic at the infrageneric level.

3.2. Chemodiversity in Salvia spp.

The genus *Salvia* consists of roughly 900 species, thus being by far the largest genus within the Lamiaceae of worldwide occurrence. *Salvia* comprises several subgenera and their sections, yet no satisfactory classification system exists (Hedge, 1982). Due to the limited number of species studied, it would seem inappropriate to correlate botanical to chemical systematics.

The quantities of exudates produced are not considerable. Compared to *Teucrium*, chemodiversity is somewhat increased. Luteolin derivatives are again higher in number than those of the apigenin series, and both series also contain 6-methoxy derivatives, exhibiting a considerable degree of methylation (Table 3). Flavonols are rarely

accumulated. All taxa listed in Table 3 have been studied for exudate constituents for the first time; however, some of the compounds have also been found as extract constituents (Hatam and Yousif, 1992; Adzet et al., 1988). For comparison, they are also listed in Table 3. Most of the published results coincide with our findings. It may thus be assumed that these compounds are also exuded. Within this genus, there is a strong tendency toward accumulation of 6-hydroxyflavones and their methyl ethers, exhibiting a wide range of chemodiversity, whereas 8-hydroxyflavones are rarely encountered. The chemodiversity among flavonols is less divergent, and notably of the 6-hydroxyflavonols, only 6-hydroxykaempferol derivatives are so far known from this genus (Lu and Foo, 2002). Interestingly, *S. arizonica* yielded no flavonoids, but instead the diterpene quinone demethylfruticulin-A (Fig. 1). This compound was earlier isolated from *S. fruticulosa* Benth., and the chemosystematic value of specific diterpenes in relation to infrageneric taxonomy was postulated (Rodriguez-Hahn et al., 1992). It would be interesting to study more *Salvia* species in this context, also with respect to the possible ecological function of diterpenoid accumulation.

3.3. Chemodiversity in Dorystoechas hastata

Dorystoechas is a monotypic genus, endemic to Turkey and belonging to the generic group around *Salvia* (Bokhari and Hedge, 1972). This assemblage also includes the genus *Perovskia* from which data on exudate flavonoids exist (Tomás-Barberán and Wollenweber, 1990). The exudate of *D. hastata* yielded only two compounds (Table 3), of which the kaempferol derivative appears to be rare within the studied group. Venturella et al. (1988) previously reported 6-OH-luteolin 6-methyl ether as a constituent in leaf extracts of another accession. In contrast, related *Perovskia* spp. yielded exudate flavonoids corresponding largely to those of *Salvia* spp. (Tomás-Barberán and Wollenweber, 1990).

3.4. Chemodiversity in Phlomis fruticosa

Phlomis, a genus consisting of some 65 species belonging to the Lamioideae, was earlier reported as being devoid of exudate flavonoids (Tomás-Barberán and Wollenweber, 1990). One accession, however, yielded small amounts of exudate consisting of apigenin and luteolin 3′-methylether. An interesting paper reported the occurrence of a number of flavone monoglycosides in hairs of *P. aurea* Decne (El-Negoumy et al., 1986), whereas the whole extracts contained corresponding diglycosides in addition. No 6-hydroxyflavonoids have been detected in any *Phlomis* spp. studied so far. Population studies of *Phlomis fruticosa* by TLC of leaf extracts were carried out earlier, but without identification of individual compounds (Margaris and Papadogianni, 1977).

4. Discussion

Most of the reports on exudate flavonoids in the Lamiaceae concern flavones with a strong tendency towards 6-substitution and/or 8-substitution. It is noteworthy that a number of taxa also accumulate 6- or 8-hydroxylated derivatives, thus differing from other families such as the Asteraceae, where 6-methoxylated derivatives mainly occur (Wollenweber and Valant-Vetschera, 1996). Earlier surveys suggested that 6- versus 8-substitution may be specific at the generic level in Lamiaceae (Tomás-Barberán and Wollenweber, 1990). Aglycone profiles also appeared to be relevant at the infrageneric level (Hernandez et al., 1987; Harborne et al., 1986; Barberán et al., 1985). Although the degree of chemodiversity is quite high within the family, the species studied here reflect that characteristic only weakly. In comparison to the Asteraceae (Wollenweber et al., 1996), however, the Lamiaceae exhibit less diverse substitution patterns, particularly with the flavonols, and thus in this case a lower degree of chemodiversity.

As for the accumulation site of exudate flavonoids, reference is made to the histochemical and phytochemical study of *Salvia blepharophylla* Brandegee ex Epling (Bisio et al., 1999). This species bears both peltate and capitate glandular hairs on its leaf surface, a common feature of the Lamiaceae (Werker et al., 1985).

Peltate hairs, being fully developed in mature leaves, release their secretions only upon being touched. In contrast, secretory material is actively extruded by the two different types of capitate hairs, which are fully active in young leaves. The presence of polyphenolic compounds was mainly observed in peltate hairs. It appears that essential oils produced by these different types of hairs are composed of different terpenoid derivatives (Werker et al., 1985). Histochemical studies in *Salvia* also indicated different qualities of products accumulated in the three types of hairs (Bisio et al., 1999). Within the Lamiaceae, more anatomical distribution studies of hair types and their relation to secondary product accumulation are highly needed to better understand these plant defense strategies. This should facilitate the interpretation of exuded flavonoid accumulation, particularly concerning incoherent patterns within or between taxa.

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